DETERMINATION OF LEAD-210 IN URANIUM BEARING REFERENCE MATERIALS FOR CERTIFICATION PURPOSES

FINAL REPORT

BY

MONENCO CONSULTANTS LIMITED
Acting through Monenco Analytical Laboratories

Work on this project was conducted under the auspicies of the Canada Centre for Mineral and Energy Technology, Energy, Mines and Resources, Canada.

DSS FILE NUMBER	:	26SQ.23440-3-9148-4
DSS CONTRACT SERIAL NUMBER	:	OSQ83-00166
CANMET PROJECT NUMBER	:	380101
CANMET SCIENTIFIC AUTHORITY	:	Dr. C.W. Smith

DEC 10 1984

15 pp

MICROMEDIA

TABLE OF CONTENTS

		Page
1.0	INTRODUCTION	1
2.0	ANALYTICAL PROCEDURES	1
3.0	ANALYTICAL RESULTS	2
3.0.1	Calibration	2
3.0.2	Lead-210 Contents Of The Standard Ores	4

APPENDIX A — AMERSHAM RBZ.44 STANDARD SOLUTION

APPENDIX B — RAW DATA USED FOR CALCULATION OF ^{210}Pb CONTENT OF ORES

TABLES

TABLE I	STANDARD CALIBRATION SOLUTION	2
TABLE II	CALCULATION OF CALIBRATION FACTOR FOR BETA COUNTER	3
TABLE III	SUMMARY OF ANALYSIS - ^{210}Pb	5

1.0 INTRODUCTION

This work was undertaken to measure the Lead-210 activity contents in each of four Canadian Certified Reference Materials Project (CCRMP) uranium and uranium-thorium ore materials. The materials, identified as BL-5, BL-4a, DH-1a and DL-1a were analysed by Dr. N. Chiu and Dr. J. Dean using gross beta counting procedures. The methodology is described below.

2.0 ANALYTICAL PROCEDURES

All samples were analysed as received. That is, the moisture contents of each sample were considered to be negligible. For three of the samples, BL-4a, DH-1a and DL-1a, analyses were carried out on at least 0.5 gram (weighed to ±0.001gram) aliquots of each material. For BL-5, 0.5 gram aliquots were digested, diluted to 100 ml and then a 10 ml. aliquot was taken. Each material was analysed 4 times. All samples were put into solution by $HF/HNO_3/HClO_4$ digestion in teflon beakers. After removal of the acids the residue was taken up in 2N HCl. The 5.01 day half-life daughter, ^{210}Bi, of ^{210}Pb was then extracted from solution with .1% DDTC (diethylammonium diethyldithiocarbonate) in chloroform. The ^{210}Bi was carried by coprecipitation with bismuth oxychloride, BiOCl and beta counted. The ^{210}Pb was then calculated from the ^{210}Bi activity.

The DDTC extractions were done in batches of 6 and the calibration was carried out using Amersham RBZ.44 solution SO/8/54 provided by Canmet. A total of 32 determinations were carried out.

The ^{210}Bi carried by BiOCl was beta counted for a length of time depending on the level of activity. Each counting period was broken up into 30 minute segments a[illegible] the counts obtained using that segment were recorded. Correc[illegible] for ^{210}Bi decay were then made on each segment and then totali[illegible] ether at the completion of the counting period. Our aim was to ob[illegible]n counting precisions of at least 1%.

3.0 ANALYTICAL RESULTS

3.0.1 Calibration

The calibration factor for the beta particle counter was determined from sources prepared using the solution of Amersham RBZ.44 supplied by Canmet. The solution was calculated to have an activity of 681 pCi ml^{-1} (25.2 BQ ml^{-1}) as of January 15, 1984. Table I contains the calculations used to obtain that activity. The available information on the standard is included in Appendix A.

The data used to calculate the calibration factor are included in Table II. The average calibration factor was found to be 0.414±0.010 (1 σ) cpm/pCi (11.19 cpm/BQ).

TABLE I - Standard Calibration Solution

Solution was known to contain 684 pCi.ml^{-1} ^{210}Pb as of December 1, 1983. As of January 15, 1984 the solutions contains:

$$684\ e^{\frac{-0.693 \times 1.5}{22 \times 12}} = 681.\ pCi.ml^{-1}\ (25.20\ BQ.ml^{-1})$$

TABLE II - Calculation of Calibration Factor For Beta Counter

	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)
Background Counts	135.	135.	135.	135.	135.	135.	135.	135.	135.	135.	135.	135.
Counting Time, Minutes	80.	80.	80.	80.	80.	80.	80.	80.	80.	80.	80.	80.
CPM	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7	1.7
Gross Sample Counts*	1747.	44,393.	32,980.	33,392.	33,580.	25,403.	33,775.	17,763.	45,809.	35,006.	34,151.	38,823.
Counting Time, Minutes	30.	163.	120.	120.	120.	30.	30.	120.	80.	120.	120.	66.7
^{210}Pb Activity, pCi	136.3	681.	681.	681.	681.	2,043.	2,724.	340.5	1,362.	681.	681.	1,362.
Calibration Factor, cpm/pCi	0.415	0.397	0.401	0.406	0.408	0.414	0.413	0.430	0.419	0.426	0.415	0.426
Precision, 1σ, cpm/pCi	0.010	0.002	0.002	0.002	0.002	0.003	0.002	0.004	0.002	0.003	0.003	0.003

*corrected for decay of ^{210}Bi

Average Calibration Factor = 0.414±0.010 (1σ)

3.02 Lead-210 Contents Of The Standard Ores

Table III shows a summary of the lead-210 contents determined in each subsample of the four standards. The data are expressed in units of pci/g and in units of BQ/g. The components used to calculate the precision of each measurement are included in Appendix B.

An illustration of the calculation used to calculate the ^{210}Pb content of sample DL-1a is as follows:

$$^{210}Pb,\ BQ/g = \frac{(23{,}623 - 1.7 \times 690)}{690 \times 0.414 \times 2.003 \times 27.027} = 1.452$$

$$\sigma^{210}Pb = \left[\left[\frac{(23{,}623 + 1.7 \times 690)^{\frac{1}{2}}}{(23{,}623 - 1.7 \times 690)}\right]^2 + \left[\frac{.010}{0.414}\right]^2\right]^{\frac{1}{2}} \times 1.452$$

$$= \left[4.91 \times 10^{-5} + 58.0 \times 10^{-5}\right]^{\frac{1}{2}} \times 1.452$$

$$\sigma^{210}Pb = 0.036$$

TABLE III

SUMMARY OF ANALYSIS - ^{210}Pb

Sample Description (Run No.)		pCi/g	BQ/g
DL-1a	(I)	39.23± 0.96	1.452±0.036
	(II)	36.90± 0.90	1.366±0.034
	(III)	38.30± 0.93	1.418±0.035
	(IV)	38.19± 0.93	1.414±0.035
	Average (n=4)	38.2 ± 1.0	1.41 ±0.04
DH-1a	(I)	809. ±20.	29.94 ±0.73
	(II)	806. ±20.	29.83 ±0.73
	(III)	754. ±18.	27.90 ±0.68
	(IV)	719. ±18.	26.59 ±0.68
	Average (n=4)	772. ±44.	28.6 ±1.6
BL-4a	(I)	406.7 ± 9.8	15.05 ±0.36
	(II)	407.2 ± 9.8	15.06 ±0.36
	(III)	380.2 ± 9.2	14.07 ±0.33
	(IV)	410.3 ± 9.9	15.18 ±0.37
	Average (n=4)	401. ±14.	14.84 ±0.53
BL-5	(I)	22,669. ±555.	839. ±20.
	(II)	22,638. ±555.	838. ±20.
	(III)	22,051. ±540.	816. ±20.
	(IV)	22,646. ±555	838. ±20.
	Average (n=4)	22,501. ±300.	833. ±11.

APPENDIX A

Amersham RBZ.44 Standard Solution

CCRMP LEAD-210 REFERENCE SOLUTION

Source material: Amersham RBZ.44, solution SO/8/54, certificate P18537 (attached).

Dilution description:

A diluent solution containing 0.18 M nitric acid, and 100 µg per mL of lead, and 99 µg per mL of bismuth was prepared from an atomic absorption standard solution of lead in 1% nitric acid, high-purity bismuth metal (6-9 s) dissolved in nitric acid, and reagent-grade nitric acid. The source material was transferred quantitatively to a 100 mL volumetric flask by twelve successive rinsing of the vial with the diluent. This solution was diluted to the mark with the diluent and mixed thoroughly. After three days, 14.98±0.02 mL of this stock was transferred to a 1L volumetric flask, and the solution was diluted to the mark with the diluent and mixed thoroughly. Aliquots of 49.85±0.03 mL of this solution were transferred to 125 mL Nalgene bottles, which were capped and sealed for shipping (831120).

Activity calculation:

The calculated activity of ^{210}Pb at 831201 is 25.3 Bq mL^{-1} (684 pCi mL^{-1}), based on the certified activity, a decay period of 3.584 y, a half-life of 22.3 y, and the dilution. The total activity uncertainty is estimated as ±2.8% (3σ = 0.6%, systematic error in activity = 1.8%, and estimated maximum systematic error in dilution = 0.4%).

Matrix:

The solution is 0.18 M in nitric acid and contains 100 µg/mL Pb and 99 µg/mL Bi.

Total activity:

Based on 49.85 mL of solution at 831201:

total activity of ^{210}Pb	=	1261 Bq or 0.0341 µCi	(2.8%)
total activity of ^{210}Bi	=	1262 Bq or 0.0341 µCi	
total activity of ^{210}Po*	=	1284 Bq or 0.0347 µCi	
total activity of radio-isotopes	=	3807 Bq or 0.103 µCi	

*Assumes no plate-out.

USERS SHOULD CONFIRM ABOVE DATA AND CALCULATIONS TO THEIR SATISFACTION, AND UPDATE ACTIVITIES TO MEASUREMENT TIMES.

ENQUIRIES: C.W. SMITH (613) 995-4316

Amersham UK

[redacted]

Description

Principal radionuclide: Lead-210 — Product code: RBZ.44

Daughter radionuclide: Bismuth-210 + Polonium-210 — Solution number: S0/8/54

Measurement

Reference time:	1200 GMT on 1 May 1980	
Radioactive concentration of lead-210:	0.958	microcuries per gram of solution
which is equivalent to:	35.4	kilobecquerels per gram of solution
Mass of solution:	5.3303	grams
Total activity of lead-210:	5.10	microcuries
which is equivalent to:	189	kilobecquerels

Method of measurement:
The solution was standardized by repeated 4 pi alpha counting over a period of 82 days.

Accuracy

Overall uncertainty in the radioactive concentration quoted above: ± 2.4 %

Random uncertainty: ± 0.6 % Systematic uncertainty: ± 1.8 %

Overall uncertainty is defined on the reverse of the certificate.

Radionuclidic Purity

The estimated activities of any radioactive impurities found by high resolution gamma ray spectrometry, or in any other examination of the solution, are listed below expressed as percentages of the activity of the principal radionuclide at the reference time:

Other radionuclides less than 0.01 %

Chemical Composition

2M HNO_3 containing 100 micrograms of lead and 100 micrograms of bismuth per ml.

Physical Data

Recommended half-life: 22.3 ± 0.2 years

Lead-210:	100% beta particle emission.	
Bismuth-210:	100% beta particle emission.	Half-life 5.01 ± 0.01 days
Polonium-210:	100% alpha particle emission.	Half-life 138.38 ± 0.01 days

Remarks

The ratio of the activity of of bismuth-210 to the activity of lead-210 = 1.0006
The ratio of the activity of of polonium-210 to the activity of lead-210 = 0.9559 at the reference time and will slowly increase to a theoretical maximum of 1.0179

Tests made over 4 years on standardized solutions of lead-210 stored in glass ampoules have shown that loss of lead-210 from solution is negligible other than by radioactive decay. There is no guarantee that all the polonium-210 will remain in solution.

Approved

Physics Department

for Managing Director

Methods of measurement

The measurement techniques listed below are currently in use at the Radiochemical Centre for the absolute standardization of radioactive solutions. The methods used for this standardization are indicated on the front of the certificate.

Using a gas flow proportional counter

A 4π beta counting
B 4π alpha counting
C 4π internal conversion electron counting
D 4π coincidence counting
E 4π anticoincidence counting
F 4π coincidence and anticoincidence counting

Using a liquid scintillation counter

G 4π coincidence counting
H 4π anticoincidence counting
J 4π coincidence and anticoincidence counting
K 4π efficiency tracing

S.I. unit of radioactivity

The S.I. unit of radioactivity is the becquerel

1 becquerel (Bq) = 1 nuclear transformation per second, therefore
1 curie (Ci) = 3.7×10^{10} becquerels exactly.

Useful conversion factors are:

1 microcurie (μCi) = 3.7×10^{4} Bq = 37 kilobecquerels (kBq)
1 millicurie (mCi) = 3.7×10^{7} Bq = 37 megabecquerels (MBq)
1 kilobecquerel (kBq) = 27.027 nanocuries (nCi)
1 megabecquerel (MBq) = 27.027 microcuries (μCi)

Overall Uncertainty

The overall uncertainty was calculated in accordance with the recommendations of the International Commission on Radiation Units and Measurements (ICRU Report 12). The limits of uncertainty were taken as the arithmetic sum of the uncertainty due to random variations, calculated at the 99.7% confidence level, and the estimated systematic uncertainties in the measurement.

Lead-210 Reference Solution

Packaging Description

- 50ml of a 0.1M nitric acid solution containing ca. 0.039 μCi of each of ^{210}Pb, ^{210}Bi, ^{210}Po is contained in a 125ml Nalgene bottle.
- The cap threads are wound with Teflon tape, and (externally) with parafilm.
- The bottle is contained in a plastic bag inside an aluminum can.

APPENDIX B

RAW DATA USED FOR CALCULATION OF ^{210}Pb CONTENT OF ORES

Measurement of Pb-210 Content of CANMET Standard Ores

Background Information

Gross Counts	135
Counting Time, Minutes	80
Counting Rate, cpm	1.7

Sample Description	DL-1a	I	II	III	IV
Gross Counts for Bi-210*		23,623	31,005	16,052	30,037
Counting Time, Minutes		690	960	480	900
Sample Weight, Grams		2.003	2.001	2.001	2.003

Sample Description	DH-1a	I	II	III	IV
Gross Counts for Bi-210*		53,833	40,432	37,702	36,054
Counting Time, Minutes		160	120	120	120
Sample Weight, Grams		0.999	1.004	1.001	1.004

Sample Description	BL-4a	I	II	III	IV
Gross Counts for Bi-210*		102,430	102,590	95,640	68,560
Counting Time, Minutes		600	600	600	400
Sample Weight, Grams		1.004	1.004	1.002	0.999

Sample Description	BL-5	I	II	III	IV
Gross Counts for Bi-210*		28,370	45,755	54,978	56,795
Counting Time, Minutes		60	96.9	120	120
Sample Weight, Grams		0.0502	0.0502	0.0500	0.0503

*corrected for decay since precip. as BiOCl

POOR

QUALITY

ORIGINAL